Halogen lamp and LED activation of resin-modified glass ionomer restorative material.  
In vitro microhardness after long term storage

ABSTRACT

The purpose of this study was to evaluate the activation of resin-modified glass ionomer restorative material (RMGI) by Knoop microhardness (KHN) in two storage conditions: 24hrs and 6 months and in two depths (0 and 2 mm).

Materials and methods

The specimens were randomly divided into 3 experimental groups (n=10) according to activation form and evaluated in depth after 24h and after 6 months of storage. Activation was performed with QTH for 40s (700 mW/cm²) and for 40 or 20 s with LED (1,200 mW/cm²). After 24 hrs and 6 months of storage at 37°C in relative humidity in lightproof container, the Knoop microhardness test was performed.

Statistics

Data were analysed by three-way ANOVA and Tukey post-tests (p<0.05).

Results

All evaluated factors showed significant differences (p<0.05). After 24 hrs there were no differences within the experimental groups. KHN at 0 mm was significantly higher than 2 mm. After 6 months, there was an increase of microhardness values for all groups, being the ones activated by LED higher than the ones activated by QTH.

Conclusion

Light-activation with LED positively influenced the KHN for RMGI evaluated after 6 months.

Keywords: Knoop microhardness; LED; Halogen light; Resin-modified glass ionomer product; Long-term storage.

Introduction

Glass ionomer is the group of materials which undergo setting through an acid-base reaction between an ion-leachable glass and a polyacid aqueous solution. The hybrid resin-modified glass ionomer restorative material (RMGI) contains resin that modifies it but maintains the glass ionomer characteristics as fluoride release [McCabe, 1998] with the addition of the mechanical behaviour of composites which are improved when compared to conventional ionomer cement [Ilie et al., 2007; Pereira et al., 2002]. A material cannot be called resin-modified glass ionomer product unless it contains a reactive ion-leachable glass and sufficient water to support an acid-base reaction [Swift Jr et al., 1995; Kakaboura et al., 1996]. The polymerisation of these materials can be produced by the acid-base chemical mechanism, with a thermal activated initiator, a photochemical mechanism through the photoinitiator activated by blue light within the range of 400-500 nm wavelength or simultaneously with both mechanisms [Cefaly et al., 2009]. Due to these characteristics, they are especially recommended for use in primary teeth [Alpoz et al., 2008].

The curing unit has to activate the camphorquinone, which is the photoinitiator commonly present in most resin composites and RMGI [Wiggins et al., 2004; Kramer et al., 2008; Stansbury, 2000; Ramp et al., 2006]. The quartz-tungsten halogen lamp provides a white light containing a spectrum of light wavelengths filtered to produce a spectral output between 400 and 500 nm [Stansbury, 2000; Ramp et al., 2006; Rueggeberg et al., 2000], and the light emitting diode (LED) curing unit provides a narrow bandwidth of light with a wavelength of 470-490 nm. The wavelength produced by LED units is closer to the camphorquinone excitation [Kramer et al., 2008; Stansbury, 2000; Ramp et al., 2006; Rueggeberg et al., 2000; Hubbezoglu et al., 2007].

There is scarce information about the influence of RMGI light activation by LED with high intensity and its behaviour after long term storage. Since Knoop hardness is an indirect method to evaluate the elastic properties, an increase in Knoop hardness of the light-cured materials over time supports the hypothesis that another reaction is occurring, which allows the materials to polymerise even after light exposure. The polyalkenoic acid-base reaction probably compensates for inadequate light activation [Alpoz et al., 2008; Roberts et al., 2009].

The purpose of this study was to evaluate the activation of a resin-modified glass ionomer restorative material (Vitremer-3M-ESPE) by halogen lamp or light-emitting diode by Knoop microhardness in two storage conditions: 24 hrs and 6 months and in two depths (0 and 2 mm). The null hypothesis tested was: microhardness of RMGI restorative material cannot be called resin-modified glass ionomer product unless it contains a reactive ion-leachable glass and sufficient water to support an acid-base reaction [Swift Jr et al., 1995; Kakaboura et al., 1996]. The polymerisation of these materials can be produced by the acid-base chemical mechanism, with a thermal activated initiator, a photochemical mechanism through the photoinitiator activated by blue light within the range of 400-500 nm wavelength or simultaneously with both mechanisms [Cefaly et al., 2009]. Due to these characteristics, they are especially recommended for use in primary teeth [Alpoz et al., 2008].

Materials and methods

A factorial design 3x2x2 with repeated measurements
was used to evaluate the variables “Photoactivation Form” in 3 levels (Table 1), “Depth” in 2 levels (Top and Bottom), and “Storage Time” in 2 levels (24 hrs and 6 months). The dependent variable evaluated was the material microhardness.

A commercial RMGI (Vitremer, 3M ESPE, St. Paul, Minnesota, USA – A3) was tested in this study (Table 2). An 11 mm diameter light tip was placed in an Optilux 501 halogen machine (Demetron/Kerr, Danbury, CT, USA) and it had its power density measured by its own radiometer, which confirmed 700 mW/cm² (± 58 mW/cm²) of irradiance. A LED source with 1,200 mW/cm² (Radii Cal - SDI Limited, Bayswater, Victoria, Australia) and 8 mm diameter light tip was used. Table 1 presents the energy density (J/cm²) calculated after multiplying the power density by the exposure time.

**Specimens preparation**

Thirty specimens were prepared according to the 3 photoactivation form groups (n=10), as presented in Table 1.

A single operator was calibrated from a pilot study and was used to prepare the specimens.

A white Teflon mold, made of two parts with a window (3 mm diameter, 2 mm high), was placed on a Mylar strip (Dentart, Polidental, São Paulo, Brazil) over a matte black background and kept stable with a metal ring. RMGI was mixed according to manufacturer’s instructions in proportion of 2:2 and inserted in single increment with a Centrix Syringe (Centrix Inc., Shelton, CT, USA). A second Mylar strip was placed on top of the uncured material. A glass slide was positioned on top of the Mylar strip and a load with 500 g was applied for 30s on top of the glass slide in order to provide a smooth surface. The load and glass slide were removed and the RMGI restorative material was irradiated. The tip of QTH or LED were clamped directly over the window filled with restorative material, to make sure that the tip of the light machine remained parallel to the surface of the RMGI restorative material (zero distance of specimen surface) [Shortall et al., 2005].

The Teflon mold was removed from the metallic ring and the Mylar strips were discarded. Each disc of resin composite was removed from the mold and was individually stored in a light-proof container for 24 hrs and 6 months in relative humidity at 37ºC.

**Knoop microhardness**

Using a microhardness tester (PanTec, Panambra Ind. e Técnica SA, São Paulo, Brazil), a 25 g load was applied for 20 s with a dwell time of 15 s. Ten randomized indentations (5 on the top, 0 mm, and 5 on the bottom

<table>
<thead>
<tr>
<th>Storage time</th>
<th>Depth</th>
<th>Groups</th>
<th>Light source and activation time</th>
<th>Energy density (J/cm²)</th>
<th>N</th>
</tr>
</thead>
<tbody>
<tr>
<td>24hrs</td>
<td>0mm</td>
<td>TOP</td>
<td>G1 QTH (700mW/cm²) - 40s</td>
<td>28</td>
<td>10</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>G2 LED (1,200mW) - 40s</td>
<td>48</td>
<td>10</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>G3 LED (1,200mW) - 20s</td>
<td>24</td>
<td>10</td>
</tr>
<tr>
<td></td>
<td>2mm</td>
<td>BOTTOM</td>
<td>G4 QTH (700mW/cm²) - 40s</td>
<td>28</td>
<td>10</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>G5 LED (1,200mW) - 40s</td>
<td>48</td>
<td>10</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>G6 LED (1,200mW) - 20s</td>
<td>24</td>
<td>10</td>
</tr>
<tr>
<td>6 months</td>
<td>0mm</td>
<td>TOP</td>
<td>G7 QTH (700mW/cm²) - 40s</td>
<td>28</td>
<td>10</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>G8 LED (1,200mW) - 40s</td>
<td>48</td>
<td>10</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>G9 LED (1,200mW) - 20s</td>
<td>24</td>
<td>10</td>
</tr>
<tr>
<td></td>
<td>2mm</td>
<td>BOTTOM</td>
<td>G10 QTH (700mW/cm²) - 40s</td>
<td>28</td>
<td>10</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>G11 LED (1,200mW) - 40s</td>
<td>48</td>
<td>10</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>G12 LED (1,200mW) - 20s</td>
<td>24</td>
<td>10</td>
</tr>
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</table>

<table>
<thead>
<tr>
<th>Material Manufacturer</th>
<th>Composition</th>
<th>Shade</th>
<th>Lot No</th>
</tr>
</thead>
<tbody>
<tr>
<td>Vitremer</td>
<td>Powder: fluoroaluminosilicate glass, microencapsulated potassium. Persulfate, ascorbic acid and pigments</td>
<td>A3</td>
<td>8PX</td>
</tr>
<tr>
<td>3M ESPE</td>
<td>Liquid: aqueous solution of a polycarboxylic acid modified with pendant methacrylate groups, copolymer, water, HEMA and photoinitiators</td>
<td></td>
<td>8HH</td>
</tr>
</tbody>
</table>

**Table 1** - Experimental groups according to storage time, depth, light sources evaluated with irradiation times, and energy density (J/cm²) calculated.

**Table 2** - Resin-modified glass ionomer restorative material evaluated composition, shade and lot number.
surfaces, 2 mm) were made in each specimen with 100 µm distance from each other.

**Statistical analysis**

Three-way ANOVA was performed to evaluate the influence of the three variables tested: Photoactivation Form, Storage Time and Depth and the means of Knoop microhardness values (KHN) were compared by a Tukey post-test (p<0.05). The software employed was SANEST (EPAMIG, Empresa de Pesquisa Agropecuária de Minas Gerais, MG, Brazil). The software employed to calculate the sample size was Power and Precision 3 (BIOSTAT, Englewood, NJ, USA).

**Results**

Figure 2 presents the KHN mean values and standard deviation for each experimental group.
values and the Tukey's results for factor storage depth. There was a significant increase of microhardness values after 6 months storage independently of the photoactivation form or depth evaluated.

Only the interaction of Photoactivation Form and Storage Time showed statistically significant differences (p<0.003) and the results are presented in Table 4. The other double interactions between Photoactivation Form and Depth (p=0.6287); Depth and Storage Time (p=0.5427); and the triple interaction among Photoactivation Form, Depth and Storage Time (p=0.258) were not statistically significant. The microhardness values of samples activated by QTH were not significantly different from sample activated by LED after 24hs storage time. After six months, the samples showed a higher microhardness than at 24 hrs. However, the microhardness values of samples activated by QTH were significantly lower than activation by LED after 6 months storage.

Discussion

A hybrid material of glass ionomer cement and composite resin was developed to minimise the high surface wear and low physical strength of the conventional glass ionomer cements [Mount et al., 2002]. Vitremer is a hybrid material of glass ionomer restorative material and composite resin with an acid-base and photocure reaction. This type of material is reportedly set via "triple-cure" mechanisms [Swift Jr et al., 1995]. The setting reaction of Vitremer occurs with an acid-base neutralisation and free-radical methacrylate polymerisation [Swift Jr et al., 1995]. A third setting reaction was incorporated to ensure that the remaining monomer that was not affected by irradiation continued to polymerise [Mount et al., 2002]. Vitremer contains microencapsulated potassium persulphate and ascorbic acid which makes up the patented redox catalyst system that provides the methacrylate cure of the glass ionomer in the absence of light but also contains photoinitiators. It is a camphorquinone, HEMA-based restorative material [Cefaly et al., 2009]. It has been reported that the wear of the resin-modified glass ionomers is higher than that of the conventional glass ionomers, probably due to differences in matrix formation [de Gee et al., 1996].

Hse et al. [1999] stated that the resin-modified glass ionomer materials are a good alternative to amalgam for restorative procedures in children. Their popularity is related to the fluoride release and prevention of caries [Beriat and Nalbandt, 2009]. It has been reported that Vitremer exhibits an acid-base reaction after 20 min dark storage and the acid-base reaction is significantly slowed after light exposure [Kakaboura et al., 1996]. The light activation promotes a rapid polymer network formation that strongly reduces the salt formation rate [Kakaboura Jr et al., 1996].

In the present study two different activation times (40s or 20 s) of the same RMGI were evaluated because it has been reported that LED units can activate these materials in half the time [Shortall et al., 2005]. The Knoop microhardness values were not different after 24 hrs storage time reinforcing the efficiency of LED with 20s activation. Also, after 6 months storage, there were no differences in the KHN values for LED activation with 20 s or 40 s but they were higher than activation during 40 s with QTH unit. The increased microhardness after 6 months of storage may reflect the state of cure of a material and the presence of an ongoing setting reaction or maturity of the restorative materials [Basting et al., 2002]. Microhardness is important to compare the physical characteristics for dental materials. It is a parameter that reflects the behaviour of dental material in the oral environment. It has been reported that RMGI achieves the higher microhardness values after 15 days, and decreased values were recorded after 180 days storage [Ellakuria et al., 2003]. On the other hand, the same RMGI evaluated in this study presented similar values after 15 and 180 days of storage immersed in a liquid medium [Ellakuria et al., 2003] after QTH activation. The present study shows that the LED activation could increase microhardness compared with QTH activation.

It has been reported that microhardness is dependent on storage condition. The water absorption ability is related to the composition of the RMGI and the functional hydrophilic groups present in their chain. The water spreads and acts as a plastifier [Cattani-Lorente et al., 1999]. Wilde et al. reported that specimens immersed in artificial saliva of Vitremer (3M ESPE) exhibited increasing microhardness values up to the 7th day and remained stable afterwards up to the 30th day [Wilde et al., 2006]. Ellakuria et al. stated that microhardness of RMGI increased after seven days storage and was not different after 6 months storage in accordance with the present study [Ellakuria et al., 2003]. The explanation for variations on microhardness is the maturity state of a material and its setting reaction.

Mobarak et al. concluded that high intensity LED (1,100 and 1,400 mW/cm2) during 10 s achieves similar or higher microhardness values than QTH activation (350 mW/cm2 for 40 s) for Fuji II LC (GC) [Mobarak et al., 2009]. In the present study, the Knoop microhardness values of the specimens were higher at 0 than 2 mm depth, after 24 hours storage in relative humidity at 37°C. This storage condition attempted to simulate oral clinical condition. The microhardness evaluation of Cefaly et al. for Vitremer (3M ESPE) after the same storage time did not show differences for 0 or 2 mm depth but the storage condition was dry. On the other hand, Swift Jr et al. reported higher Knoop microhardness values to Vitremer (3M) at 0 mm than at 2 mm depth, in accordance with our results after 24 hrs storage in distilled water [Swift Jr et al., 1995]. By comparing the storage conditions, Cattani-Lorente et al. reported that the immersed Vitremer specimens recorded lower Vickers surface values than specimens stored at 80% < RH < 90% and similar to dry condition [Cattani-Lorente et al., 1999].

The fact that specimens activated by LED unit achieve higher Knoop microhardness values than specimens activated by QTH units in similar intensity, pointed to a favourable mechanical behaviour for this light source. In high-intensity LED units, when the photons are guided through the fiber optic, the absorbed photon energy is transformed into heat [Jeong et al., 2007]. It has been stated that LED had lower temperature increase compared
to QTH units [Price et al., 2003] but it has been reported that units with 1,000 mW/cm² intensity produce temperature increase [Price et al., 2003; Durley et al., 2008]. There is a decrease of irradiance through deep restorative materials and reflection, absorption and scattering are responsible for this [Stansbury et al., 2000; Hubbezo lu et al., 2007]. The results obtained with LED (24 or 48 J/cm²) showed that they provided the energy necessary to activate the RMGI restorative material and can be used without clinical concerns. In fact they achieve higher microhardness values than QTH activation after 6 months of storage which suggests that LED may be useful to activate RMGI material in clinical procedures.

According to the results, the null hypothesis 1 (microhardness of RMGI restorative material cured by different light-curing devices is not different) was partially accepted, the null hypothesis 2 (different time exposure with LED unit does not provide different microhardness of RMGI restorative material) was accepted, and the null hypothesis 3 was rejected (there is no difference between 24 hours and 6 months of storage for microhardness values).

Conclusion

Within the limitations of this in vitro study, it can be concluded that light-activation did not influence the Knoop microhardness values after 24 hrs of storage. There was a significant increase of microhardness values after 6 months of storage for all groups, and the ones activated by LED had higher microhardness than the ones activated by QTH. Light-activation with LED positively influenced the Knoop microhardness values for the resin-modified ionomer restorative material evaluated after 6 months of storage and the physical characteristics of these dental materials could be improved after LED activation.

Disclosure

The authors have no interest in any of the companies or products mentioned in this article.

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